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**W-8000 München 80(DE)**(54) **Water-in-oil emulsion composition.**

(57) A water-in-oil emulsion comprises (a) the oil phase of a diglyceride mixture, (b) the aqueous phase and (c) 0.01 to 10 wt.%, based the oil phase, of a phospholipid mixture comprising (c-1) a nitrogen-free phospholipid and (c-2) a nitrogen-containing phospholipid at a weight ratio of (c-1) to (c-2) of at least 1.0.

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## WATER-IN-OIL EMULSION COMPOSITION

[Field of Industrial Utilization]

This invention relates to a water-in-oil emulsion composition having excellent emulsion stability.

5

[Prior Art]

When the proportion of the aqueous phase increases in a water-in-oil emulsion composition, the emulsion state gets unstable and the separation between the aqueous phase and the oil phase and  
10 furthermore the phase inversion from the water-in-oil type to the oil-in-water type are likely to occur during the production process.

In the above emulsion composition, exudation of the moisture and oil-off occur during storage and, when bulk handling is made, destruction of emulsion and the resulting separation of the aqueous phase and rotting take place. For these reasons, this type of compositions cannot be used satisfactorily for various  
15 applications.

The use of various emulsifiers has been proposed in the past in order to solve these problems. For instance, a method which uses sucrose fatty acid esters of HLB of 1 to 4 (Japanese Patent Publication No.11014/1981), a method which uses glycerine fatty acid esters (U.S. Patent No. 3,889,005) and a method which uses polyglycerine fatty acid esters in combination with saturated fatty acid esters having a carbon  
20 number of at least 12 (Japanese Patent Laid-Open No.170432/1983) are known.

However, even when these emulsifiers are used, the problems with the water-in-oil emulsion composition described above cannot be solved sufficiently. More specifically, when the sucrose fatty acid esters of HLB of 1 to 4 or the glycerine fatty acid esters described above are used, the water-in-oil emulsion composition can be produced in fact, but the resulting product has low stability with the passage of time  
25 even when stored below normal temperatures and causes exudation of the moisture and destruction of the emulsion, followed by the separation of the aqueous phase when it is spread.

When polyglycerine ester of condensed ricinoleic acid which has particularly high emulsion stability among the polyglycerine fatty acid esters is used, the tissue of the product is soft and exhibits a paste-like form, so that shape retentivity is low and oil-off is likely to occur. In addition, when the product is kept at  
30 low temperatures, stability with the passage of time is low and the separation of the aqueous phase occurs.

As a result of various studies in search of a method of obtaining a water-in-oil emulsion composition which solves these problems, is stable with time and has high shape retentivity, the inventors of the present invention have found out first that a water-in-oil emulsion composition prepared by mixing and emulsifying an edible oil composition (oil phase) containing a predetermined amount of a diglyceride and an  
35 aqueous phase using water as the base has excellent properties in both stability and shape retentivity that could not have been observed in the conventional compositions. However, the inventors of the invention have also found out that this water-in-oil emulsion composition prepared by use of the composition containing a diglyceride alone cannot keep sufficient stability and shape retentivity during storage for an extended period of time or at high temperatures.

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( Summary of the invention )

The inventors have furthered the study in order to solve the problems described above and have  
45 recognized that the stability of the water-in-oil emulsion composition can be improved in comparison with the conventional phospholipid mixture by using a specific amount of a specific phospholipid mixture as the emulsifier of the water-in-oil emulsion composition. Furthermore, the inventors have found that when a specific amount of a diglyceride is contained in the oil phase of this water-in-oil emulsion composition, the stability of the composition can be improved drastically. The present invention has thus been completed.

50 Thus the present invention provides a water-in-oil emulsion composition having excellent stability, which is characterized by containing, as at least one component of emulsifiers, 0.01 to 10 wt% of a phospholipid mixture having a weight ratio of a nitrogen-free phospholipid to a nitrogenous phospholipid of at least 1.0 in an oil phase. Furthermore, the present invention provides a water-in-oil emulsion composition described above which contains 0.1 to 80 wt% of at least one diglyceride in the oil phase.

The water-in-oil emulsion of the invention comprises (a) the oil phase of a diglyceride mixture, (b) the

aqueous phase and (c) 0.01 to 10 wt.%, based the oil phase, of a phospholipid mixture comprising (c-1) a nitrogen-free phospholipid and (c-2) a nitrogen-containing phospholipid at a weight ratio of (c-1) to (c-2) of at least 1.0.

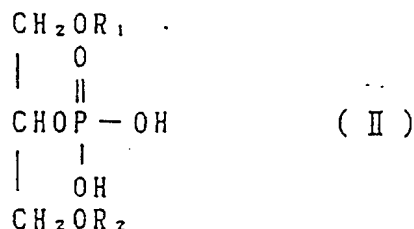
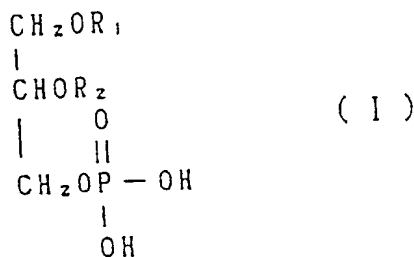
It is preferable that a weight ratio of the oil phase to the aqueous phase ranges from 99/1 to 5/95; the phospholipid mixture (c) contains at least 15 wt.%, based on the entire phospholipids, of phosphatidic acid and/or a phosphatidic acid salt; the diglyceride mixture contains 0.1 to 80 wt.% of one or more diglycerides; the diglyceride mixture contains 0.1 to 80 wt.% of one or more diglycerides and the diglycerides have fatty acid moieties of 8 to 24 carbon atoms and at least 70 wt.%, based on the entire fatty acid moieties, of unsaturated fatty acid moieties; and the phospholipid mixture (c) contains at least 15 wt.%, based on the entire phospholipids, of phosphatidic acid and/or a phosphatidic acid salt and the phosphatidic acid has the formula (I) or (II) in which R<sub>1</sub> and R<sub>2</sub> each are an aliphatic acyl group having 8 to 24 carbon atoms, saturated or unsaturated.

Now the present invention will be described in detail.

The phospholipid mixture to be used in the water-in-oil emulsion composition of the present invention is one containing a reduced amount of phosphatidylcholine or phosphatidylethanolamine wherein the weight ratio of a nitrogen-free phospholipid to a nitrogenous phospholipid is at least 1.0. The quantity of this phospholipid mixture in the water-in-oil emulsion composition of the present invention is 0.01 to 10 wt%, preferably 0.05 to 5 wt%, in the oil phase.

Examples of the phospholipid mixture containing a reduced amount of the nitrogenous phospholipid such as phosphatidylcholine or phosphatidylethanolamine which can be used in the present invention include those containing phosphatidic acid or/and its salts, phosphatidylinositol, phosphatidylglycerol and further their lysophospholipids. A particularly preferred phospholipid mixture used in the present invention is one containing at least 15 wt% of phosphatidic acid or/and phosphatidic acid salts in the total phospholipid.

The phosphatidic acid used in the present invention is one represented by the following general formula (I) and/or (II):



where R<sub>1</sub> and R<sub>2</sub> may be same or different from each other and each is a C<sub>8</sub> to C<sub>24</sub> saturated or unsaturated aliphatic acyl group.

Examples of the salts that constitute the phosphatidic acid salts are sodium, potassium, calcium, aluminum, magnesium and ammonium. The phosphatidic acid or/and phosphatidic acid salts may contain lysophosphatidic acid or/and lysophosphatidic acid salts described above.

The phospholipid mixture described above in which a weight ratio of the nitrogen-free phospholipid to the nitrogenous phospholipid is at least 1.0 can be obtained by reducing the content of phosphatidylcholine and phosphatidylethanolamine through the enzymatic treatment (treatment by phospholipase D) using natural lecithin as the starting material or through the fractionation, for example, and increasing the content of the phosphatidic acid and phosphatidylinositol above the specific values. The natural lecithin used here is one originating in vegetables such as soybean, rapeseed, sunflower, or corn, or animals such as yolk.

The phospholipid mixture described above may further be treated by fractionation with a solvent, an ion-exchange column or a silicic acid column, electrodialysis, and so forth.

Besides these phospholipids thus subjected to the enzymatic treatment, those phospholipids which are obtained by treating a monoglyceride or diglyceride with a phosphorus oxidizing agent (e.g. pentoxide, pentachloride or oxychloride) can be likewise used in the present invention.

The water-in-oil emulsion composition having excellent emulsion stability as the object of the present invention can be obtained by mixing 0.01 to 10 wt% of the resulting phospholipid mixture in the oil phase.

The emulsion stability can be further improved by mixing at least one diglyceride to the oil phase component. The diglyceride-containing fat and oil components used in the present invention can be obtained by transesterifying in the presence of an alkali metal or (and) an alkaline earth metal a mixture of glycerine with at least one fat or oil selected from the group consisting of ordinary vegetable oils such as safflower oil, olive oil, corn seed oil, rapeseed oil, coconut oil, palm kernel oil, palm oil, soybean oil, corn oil, etc., animal fats and oils such as lard, beef tallow, fish oil, milk fat, etc., or fractionated oils, transesterified oils, randomized oils and hardened oils of these vegetable and animal fats and oils, by esterifying a mixture of a fatty acid with glycerine.

In order to increase the diglyceride content of the resulting diglyceride-containing component, the treatment by molecular distillation or silicic acid column chromatography is suitable and in order to reduce the diglyceride content, on the contrary, at least one of the starting fats and oils described above may be added thereto.

The diglyceride content of the water-in-oil emulsion composition of the present invention ranges from 0.1 to 80 wt% in the oil phase, preferably from 2 to 60 wt%. It is further preferred that the diglyceride exist in liquid form in the oil phase at a temperature at which the emulsion composition is used. Preferably, the diglyceride is at least one of those consisting of  $C_2$  to  $C_{24}$ , preferably  $C_{16}$  to  $C_{22}$  fatty acid residues containing at least 70 wt% of unsaturated fatty acid residues of the total fatty acid residues.

The water-in-oil emulsion composition of the present invention may contain milk products, inorganic salts, saccharides, high-molecular polysaccharides, perfumes, acids, tasting agents, and so forth as the side components of the aqueous phase or oil phase in accordance with the intended object.

Among them, the high-molecular polysaccharides are especially effective in further improving emulsion stability at high temperatures. Particular examples of the polysaccharides include gum arabic, carrageenan, locust bean gum, xanthan gum, guar gum, tamarind seed polysaccharides, taracant gum, dextrin,  $\alpha$ -starch, starch, carboxymethylcellulose, methylcellulose, sodium alginate, and pectin.

The aqueous phase which constitutes the water-in-oil emulsion composition of the present invention and uses water as the base may consist of water alone. It may contain milk products (e.g. milk protein), perfumes, tasting agents, and the like as the side components in the same way as that described above.

In the production of the water-in-oil emulsion composition of the present invention, mixing and emulsification are preferably made so that the mixing ratio of the oil phase containing 0.01 to 10 wt% of the phospholipid mixture, wherein the weight ratio of the nitrogen-free phospholipid to the nitrogenous phospholipid is at least 1.0, to the aqueous phase is 99/1 to 5/95 by weight. It is further preferred that the mixing and emulsification are made so that 0.1 to 80 wt% of the diglyceride is contained in the oil phase.

#### [Effect of the Invention]

Since the water-in-oil emulsion composition in accordance with the present invention contains a specific amount of a specific phospholipid mixture in the oil phase, the preservation stability for a long time and high temperature resistance of the emulsion composition having a weight ratio of the oil phase to the aqueous phase of 99/1 to 5/95 can be improved drastically, and particularly the problems of exudation of the moisture, oil-off and the drop of shape retentivity that are likely to occur in the conventional water-in-oil emulsion compositions can be satisfactorily solved. Furthermore, the effect described above can be still increased by blending a specific amount of a diglyceride in the oil phase.

#### [Example]

Now the present invention will be described in further detail with reference to Reference Example, Examples and Comparative examples, though the present invention is not limited to these examples.

The terms "part(s)" and "%" in the examples are by weight unless otherwise specified.

## Reference Example 1: (Preparation of phospholipid mixture)

Soybean lecithin as the starting material was treated with phospholipase D to reduce the content of phosphatidylcholine and phosphatidylethanolamine and to increase the content of phosphatidylinositol and phosphatidic acid (and its calcium salts), thus obtaining phospholipid mixtures (1) and (2). The chemical compositions of these mixtures are given in the following Table 1.

For comparison, the chemical composition of soybean lecithin (phospholipid mixture (3)) is also given in Table 1.

Table 1:

Composition of phospholipid mixture			
Sample No.	Phospholipid mixture		
	(1)	(2)	(3)
acetone insolubles <sup>1)</sup> (%)	96.8	96.2	95.9
PC content <sup>2)</sup> (%)	0.1	0.1	29.3
PE content <sup>3)</sup> (%)	10.5	0.3	21.1
PI content <sup>4)</sup> (%)	23.1	11.2	15.8
PA content <sup>5)</sup> (%)	31.0	56.2	11.8

## Remarks:

1) acetone insolubles:

lecithin content described in "Official Standard for Food Additives"

2) PC content:

phosphatidylcholine content of the acetone insolubles (wt%); TLC analytical value in accordance with "Standard Fats and Oils Analytical Test Methods", edited by the Japan Oil Chemists' Society (2.2.8.4a.86 phospholipid phosphorus composition)

3) PE content:

phosphatidylethanolamine content of the acetone insolubles (wt%); TLC analytical value obtained by the same method as 2)

4) PC content:

phosphatidylinositol content of the acetone insolubles (wt%); TLC analytical value obtained by the same method as 2)

5) PA content:

phosphatidic acid (and its calcium salts) content of the acetone insolubles (wt%); TLC analytical value obtained by the same method as 2)

## Reference Example 2: (Production of oil composition)

After 25 parts of glycerine was added to 75 parts of refined rapeseed oil, 0.1 part of calcium hydroxide was added to the mixture to conduct transesterification in a customary manner. Then, monoglyceride was removed by molecular distillation and refining was further carried out in a customary manner to obtain a transesterified oil. The glyceride composition of this oil was 19.4% of triglyceride, 79.6% of diglyceride and 1.0% of monoglyceride.

Next, this oil was mixed with the refined rapeseed oil in the proportions specified in Table 2 to prepare oil compositions (2), (3), (4) and (5). The oil composition consisting of the refined rapeseed oil alone was referred to as the oil composition (1).

Oil compositions (6) and (7) were prepared in the same way by using palm oil and lard as the starting materials, respectively.

The glyceride compositions of these compositions are given in Table 2.

Table 2:

Glyceride composition of diglyceride-containing oil compositions (%)							
Oil composition	(1)	(2)	(3)	(4)	(5)	(6)	(7)
Oil source	refined rapeseed oil	refined rapeseed oil	refined rapeseed oil	refined rapeseed oil	refined rapeseed oil	palm oil	lard
Blend ratio of transesterified oil to refined rapeseed oil, palm oil or lard (weight ratio)	0/100	6/94	13/87	25/75	38/62	38/62	36/64
Glyceride composition <sup>1)</sup> (%)							
triglyceride	98.7	94	88	79	69	68	70
diglyceride	1.2	5	11	20	30	30	27
monoglyceride	0.1	1	1	1	1	2	3
Remarks:							

1) The glyceride composition (%) was determined by gas chromatography.

#### Examples 1 and 2:

Oil phases were prepared by blending 0.05, 0.1, 0.2, 0.5 and 1.0 wt%, based on the refined rapeseed oil, of the phospholipid composition (1) or (2) prepared above with 50 parts of the refined rapeseed oil and 50 parts of distilled water was added to each of these oil phases. Each mixture was emulsified with a homomixer to prepare a water-in-oil emulsion composition.

Each of the resulting compositions was transferred into a graduated test tube and either left settled at 25 °C for 24 hours or at 80 °C for 30 minutes, or centrifuged at 3000 rpm for 5 minutes after being left settled at 80 °C for 30 minutes. The proportion of the separated aqueous phase to the total aqueous phase was measured so as to evaluate emulsion stability. The results are given in Table 3.

#### Comparative Example 1:

Oil phases were prepared by blending 0.05, 0.1, 0.2, 0.5 and 1.0 wt%, based on the refined rapeseed oil, of the phospholipid mixture (3) to 50 parts of the refined rapeseed oil and 50 parts of distilled water was added to each oil phase. Each mixture was emulsified with a homomixer to obtain a water-in-oil emulsion composition.

The emulsion stability of each of these emulsion compositions was evaluated by the same method as that of Examples 1 and 2. The results are given in Table 3.

Table 3:

Stability evaluation of emulsion					
	Kind of phospholipid mixture blended	Quantity of phospholipid mixture (based on oil; wt%)	Emulsion stability <sup>1)</sup>		
			25 °C, 24 hr	80 °C 30 min	centrifugal separation at 3000 rpm for 5 min after settled at 80 °C for 30 min
Example 1	phospholipid mixture (1)	0.05	9	18	35
		0.1	0	7	18
		0.2	0	0	0
		0.5	0	0	0
		1.0	0	0	0
Example 2	phospholipid mixture (2)	0.05	0	0	0
		0.1	0	0	0
		0.2	0	0	0
		0.5	0	0	0
		1.0	0	0	0
Comparative Example 1	phospholipid mixture (3)	0.05	88	92	96
		0.1	70	90	96
		0.2	48	85	90
		0.5	20	76	84
		1.0	7	72	77

Note:

1) Each emulsion composition prepared was either settled at 15 °C for 24 hours and at 80 °C for 30 minutes or settled at 80 °C for 30 minutes and then centrifuged at 3000 rpm for 5 minutes, and the proportion (vol%) of the separated aqueous phase to the total aqueous phase was employed to represent stability.

Example 3 to 6:

Oil phases were prepared by blending 0.1 wt% of the phospholipid mixture (1) listed in Table 1 into 50 parts of each of the oil compositions (1) to (4) listed in Table 2, and 50 parts of distilled water was added to each of the oil phases. Each mixture was emulsified with a homomixer to prepare a water-in-oil emulsion composition.

The emulsion stability of each of the resulting emulsion compositions was evaluated by the same method as that of Examples 1 and 2. The results are given in Table 4.

Comparative Examples 2 to 5:

Oil phases were prepared by blending 0.1 wt%, based on each oil compositions (1) to (4) listed in Table 2, of the phospholipid mixture (3) listed in Table 1 with 50 parts of the oil compositions (1) to (4), and 50 parts of distilled water was added to each of the oil phases. Each mixture was emulsified with a homomixer to prepare a water-in-oil emulsion composition.

The emulsion stability of each of these emulsion compositions was evaluated by the same method as that of Examples 1 and 2. The results are given in Table 4.

Table 4

		Kind of oil composition	Kind of phospholipid mixture	Emulsion stability		
				25 °C, 24 hr	80 °C 30 min	centrifugal separation at 3000 rpm for 5 min after settled at 80 °C for 30 min
Example	3	(1)	phospholipid mixture (1)	0	7	18
	4	(2)		0	0	0
	5	(3)		0	0	0
	6	(4)		0	0	0
Comparative Example	2	(1)	phospholipid mixture (3)	70	90	96
	3	(2)		13	85	95
	4	(3)		0	73	92
	5	(4)		0	20	48

Example 7

A water-in-oil emulsion was prepared by mixing 0.1 wt%, based on the oil composition, of the phospholipid mixture (1) shown in Table 1 with 40 parts of the oil composition (4) shown in Table 2 to prepare an oil phase and then mixing the oil phase with 60 parts of distilled water, emulsified with a homomixer.



## Example 8

A water-in-oil emulsion was prepared by mixing 0.1 wt%, based on the oil composition, of the phospholipid mixture (1) shown in Table 1 with 30 parts of the oil composition (4) shown in Table 2 to prepare an oil phase and then mixing the oil phase with 70 parts of distilled water, emulsified with a homomixer.

## Comparative Example 6

A water-in-oil emulsion was prepared by mixing 0.1 wt%, based on the oil composition, of the phospholipid mixture (3) shown in Table 1 with 40 parts of the oil composition (4) shown in Table 2 to prepare an oil phase and then mixing the oil phase with 60 parts of distilled water, emulsified with a homomixer.

## Comparative Example 7

A water-in-oil emulsion was prepared by mixing 0.1 wt%, based on the oil composition, of the phospholipid mixture (3) shown in Table 1 with 30 parts of the oil composition (4) shown in Table 2 to prepare an oil phase and then mixing the oil phase with 70 parts of distilled water, emulsified with a homomixer.

The emulsion compositions obtained in Examples 7 and 8, Comparative Examples 6 and 7 were evaluated in view of the emulsion stability by the same method as that of Example 1. Results are shown in Table 5, which evidently show the water-in-oil emulsion compositions obtained in Examples are stabler as emulsions having a large amount of water. In the table, figures in the rightmost column show percent by volume of the separated aqueous phase to the total aqueous phase after the centrifugal separation.

Table 5

		Kind of oil composition	Kind of phospholipid mixture	Ratio of aqueous phase to oil phase	Emulsion stability
					Centrifugal separation at 3000 rpm for 5 min after settled at 80 °C for 30 min
Example	6	(4)	(1)	50/50	0
	7			60/40	0
	8			70/30	0
Comparative Example	5	(4)	(3)	50/50	48
	6			60/40	85
	7			70/30	89

## Claims

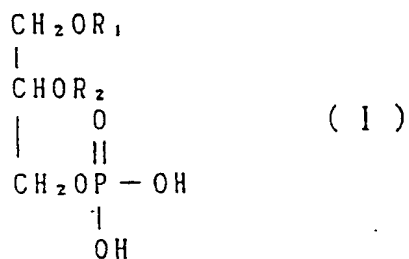
1. A water-in-oil emulsion comprising (a) the oil phase of a diglyceride mixture, (b) the aqueous phase and (c) 0.01 to 10 wt.%, based the oil phase, of a phospholipid mixture comprising (c-1) a nitrogen-free phospholipid and (c-2) a nitrogen-containing phospholipid at a weight ratio of (c-1) to (c-2) of at least 1.0.
2. The emulsion as claimed in Claim 1, in which a weight ratio of the oil phase to the aqueous phase ranges from 99/1 to 5/95.
3. The emulsion as claimed in Claim 1, in which the phospholipid mixture (c) contains at least 15 wt.%, based on the entire phospholipids, of phosphatidic acid and/or a phosphatidic acid salt.
4. The emulsion as claimed in Claim 1, in which the diglyceride mixture contains 0.1 to 80 wt.% of one or

more diglycerides.

5. The emulsion as claimed in Claim 1, in which the diglyceride mixture contains 0.1 to 80 wt.% of one or more diglycerides and the diglycerides have fatty acid moieties of 8 to 24 carbon atoms and at least 70 wt.%, based on the entire fatty acid moieties, of unsaturated fatty acid moieties.

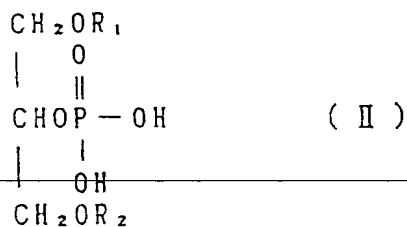
6. The emulsion as claimed in Claim 1, in which the phospholipid mixture (c) contains at least 15 wt.%, based on the entire phospholipids, of phosphatidic acid and/or a phosphatidic acid salt and the phosphatidic acid has the formula (I) or (II) in which R1 and R2 each are an aliphatic acyl group having 8 to 24 carbon atoms, saturated or unsaturated.

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## EUROPEAN SEARCH REPORT

Application Number

EP 90 11 6677

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
P,X,L	EP-A-0 378 893 (KAO CORP.) * Complete * Document quoted "L" for its' casting doubt on the "first deposit" character of JP 226842/89 and thus validity of the convention priority claim - - - -	1-6	A 23 D 9/00 A 23 J 7/00
Y	EP-A-0 171 112 (UNILEVER N.V.) * Complete * - - - -	1,2,4,5	
Y	EP-A-0 075 463 (KYOWA HAKKO KOGYO K.K.) * Abstract; page 1, line 8 - page 2, lines 16,35 - page 12, line 28; examples 1-3; claims 9,10 * - - - -	1,2,4,5	
Y	US-A-2 062 782 (A.K. EPSTEIN et al.) * Complete * - - - -	1,2,4,5	
Y	US-A-2 089 470 (A.K. EPSTEIN et al.) * Complete * - - - -	1,2,4,5	
Y	US-E-2 890 3 (R. ANEJA et al.) * Abstract; column 1, line 10 - column 4, line 2; column 6, lines 4-8; examples 1,3-7 * - - - -	1,2,4,5	
Y	US-A-3 896 975 (D.W. FOLLMER) * Column 2, lines 3-15,29 - column 3, line 49; column 5, line 58 - column 7, line 42; examples 1,3; claims 1,4,5,17 * - - - - -	1,2,4,5	
The present search report has been drawn up for all claims			
Place of search		Date of completion of search	Examiner
The Hague		12 December 90	FISCHER W.H.F.
<div>CATEGORY OF CITED DOCUMENTS</div> <div>X : particularly relevant if taken alone</div> <div>Y : particularly relevant if combined with another document of the same category</div> <div>A : technological background</div> <div>O : non-written disclosure</div> <div>P : intermediate document</div> <div>T : theory or principle underlying the invention</div> <div>E : earlier patent document, but published on, or after the filing date</div> <div>D : document cited in the application</div> <div>L : document cited for other reasons</div> <div>&amp; : member of the same patent family, corresponding document</div>			



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